EFFECT OF SHAPE SIZE AND CONTENT ON THE EFFECTIVE THERMAL CONDUCTIVITY BeO FILLED POLYMER COMPOSITES

A Project Report Submitted in Partial Fulfillment of the Requirements for the Degree of

B. Tech.

(Mechanical Engineering)

By

ABHISHEK PANDA

Roll No. 109ME0398

Department of Mechanical Engineering

NATIONAL INSTITUTE OF TECHNOLOGY

ROURKELA

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Rourkela

C E R T I F I C A T E

This is to certify that the work in this thesis entitled **Effect of Shape , Size and Content on the Effective Thermal Conductivity of BeO Filled Polymer Composites** by **Abhishek Panda,** has been carried out under my supervision in partial Fulfilment of the requirements for the degree of **Bachelor of Technology** in *Mechanical Engineering* during session 2012 - 2013 in the Department of Mechanical Engineering, National Institute of Technology, Rourkela.

To the best of my knowledge, this work has not been submitted to any other University/Institute for the award of any degree or diploma.

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(A)

ABSTRACT

Particulate filled polymeric composites with enhanced thermo-physical properties are highly in demand in electronic industry. This project presents a numerical and analytical investigation on the thermal conductivity enhancement of Beryllium oxide filled polymer composites

In the numerical study, the finite-element package ANSYS is used to calculate the conductivity of the composites. Three-dimensional cube-in-cube and sphere-in-cube lattice array models are used to simulate the microstructure of composite materials for various filler size and concentration. This study reveals that the incorporation of Beryllium oxide particulates results in improvement of thermal conductivity of polymer resin. The experimentally measured conductivity values are compared with the numerically calculated ones and it is found that the values obtained for various composite models using finite element method (FEM) are in reasonable agreement with the experimental values.

Key Words: *Polymer Composite, polymer resin, Beryllium oxide, thermal Conductivity, FEM.*

(B)

LIST OF TABLES AND FIGURES

Chapter 1
Introduction

1. INTRODUCTION

Composites or composite materials are available in nature or engineered fusing two or more materials with considerably different chemical and physical properties which remain distinct at microscopic or macroscopic level within the finished structure. The constituent material is basically of two categories: reinforcement and matrix, the matrix supports the reinforcement against mechanical and environmental damage by surrounding and maintaining their relative position, while the reinforcement bestow physical properties and special mechanical such as dielectric, strength, stiffness etc.

Metallic oxides and metals in epoxy and other resins prove to be quite effective in providing those characteristics as they (the composites) have good thermal conductivity of the fillers thus increasing their applications in the field of electronics

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TYPES OF COMPOSITE MATERIALS:

Basically, composites can be categorized into three groups on the basis of matrix material. They are:

a) Metal Matrix Composites (MMC)

b) Ceramic Matrix Composites (CMC)

c) Polymer Matrix Composites (PMC)

a) Metal Matrix Composites:

Metal matrix composites have many advantages over monolithic metals like higher specific modulus, higher specific strength, better properties at elevated temperatures and lower coefficient of thermal expansion. Due to these characteristics metal matrix composites are considered for wide range of applications like combustion chamber nozzle (in rocket, space shuttle), cables, housings, tubing, structural members, heat exchangers etc.

b) Ceramic matrix Composites:

The main reason behind producing ceramic matrix composites is to increase the toughness. Obviously, it is hoped and indeed often found that there is a concomitant improvement in strength and stiffness of ceramic matrix composites.

c) Polymer Matrix Composites:

These are the most abundantly used matrix material. Generally the mechanical properties of polymers are inadequate for many structural purposes, particularly their low strength and stiffness as compared to metals and ceramics. These difficulties are overcome by reinforcing other materials with polymers. Secondly processing of this type of matrix composites does not demand high pressure and high temperature. Simpler equipments are required for manufacturing polymer matrix composites. For this reason polymer composites developed rapidly and soon became popular for structural applications. Polymer composites are used because overall properties of these composites are superior to those of the individual polymers. The elastic modulus is greater than that of the neat polymer but is not as brittle as ceramics.

Polymer composites can be classified into following three groups on the basis of reinforcing material. They are:

(a) Fibre reinforced polymer (FRP)

(b) Particle reinforced polymer (PRP)

(c) Structural polymer composites (SPC)

In the study conducted by Sonam Agarwal [9] on TiO2 in epoxy it was found that the thermal conductivity increased quite significantly allowing it to be used in electronic packaging, encapsulations, die (chip) attach, thermal grease, thermal interface material and electrical cable insulation. Richard.F.Hill and Peter H. Supancic [2] conducted a study by comparing thermal and mechanical characteristics of BN ($PolarTherm^{TM}$, Grade PT 110, Advanced Ceramics Corp., Cleveland, OH),α-phase SiC (Third Millennium Products, Knoxville, TN), TiB2 (Grade HCT-30, Advanced Ceramics

Corp.), and α-phase Al_2O_3 in epoxy. He found that BN had some significance over the other materials in terms of thermal conductivity.

Beryllium oxide was hence chosen as the material for filler owing to its high thermal which would facilitate its use as heat sinks in electronic devices.

Chapter 2

Literature Review

2. LITERATURE REVIEW

In 1988 P. Bujard [1] prepared castable particulate filled epoxy resins exhibiting excellent thermal conductivity using hexagonal boron nitride as filler. He observed that the thermal conductivity of boron nitride filled epoxies is influenced by the sample preparation procedures due to the agglomeration effects of the particles in the matrix. He measured the temperature dependence of thermal conductivity of resins as a function of volume content of filler. He also found that the thermal conductivity in percolative systems depends in a complex way on the filler concentration and temperature. In that paper he discussed the mechanism leading to the observed behaviour. Hill and Supanic [2] used platelet-shaped particles of similar size and shape and investigated them as fillers for improving the thermal conductivity of polymer–ceramic composite materials. They found that the conductivities of composites filled with hard, stiff ceramic particles exceeded 3.5 W/(mK), or >20 times the conductivity of the polymer matrix, those showed to be almost independent of the intrinsic filler conductivity range of 33–300 W/(mK). On the contrary, the thermal conductivity of composites filled with soft, platelet-shaped BN fillers reached over 13 W/(mK). Hence they proposed a mechanism whereby deformation of the soft filler particles provided improved particle-to-particle connectivity and allowed greater packing density thus resulting in the ability to achieve higher conductivity than is possible for hard and stiff particles of similar morphology. Their experimental results discussed took into account various thermal conductivity prediction models in the literature. In 2004, Lianhua Fan et.al. [3] highlighted that composites as such have unique characteristics combining the low-temperature processability of organic polymer matrix and the various functionalities endowed by the other component in the composites. Furthermore, electrically conductive adhesives (ECAs) had been explored as an environment friendly interconnection technique. Along with the many potential advantages for surface mount and flip chip applications, typical ECA materials suffered from a few critical issues that limited its use as a drop-in replacement for lead-containing solders. Making an attempt to understand and improve the thermo-mechanical properties of ECA materials, they introduced nano-sized silver particles into the conventional ECA compositions. The nano particles influence on bulk resistivity has been reported in this paper, as maintaining an acceptable conductivity which is imperative for high performance and environmentally benign interconnections. They found that the bulk resistivity of ECA formulations strongly depended on the contents of nano particles and silver flake, as well as the surface properties and particle morphology. Thermal conductivity of alumina based composite samples was also affected upon the inclusion of nano alumina particles. The electrical and thermal conductivities of the polymer composites containing nano particles were determined by the contacts of micro-sized particles and interfaces that involved nano particles along the conduction paths. Wenying et.al.[4] in 2007 published their work on the thermal conductivity of boron nitride (BN) particulates reinforced high density polyethylene (HDPE) composites. They investigated the composite under a special dispersion state of BN particles in HDPE, i.e., BN particles surrounding HDPE particles. The effects of particulate content, particle size of HDPE and temperature on the thermal conductivity of the composites were discussed. Results showed that the special dispersion of BN in matrix provides the composites with high thermal conductivity; moreover, the thermal conductivity of composites is higher for the larger size HDPE than for the smaller size one. Increasing filler content increases the thermal conductivity, and the results significantly deviate from the theoretic models. It was also found that the combined use of BN particles and alumina short fibre gave higher thermal conductivity of composites compared to the BN particles used alone. Rajinder Pal [5] in 2007 Pal developed three new models for the effective thermal conductivity of concentrated particulate composites using the differential effective medium approach. One model predicted the relative thermal conductivity (Kr) of a particulate composite to be a function of two variables, namely thermal conductivity ratio λ (ratio of dispersed-phase to continuous-phase conductivities) and volume fraction of particles . The other two models predict Kr to be a function of three variables, namely, λ, ϕ, and ϕm where ϕm is the maximum packing volume fraction of particles. The proposed models are evaluated using twelve sets of published experimental data on the thermal conductivity of particulate composites, covering broad ranges of λ and ϕ.Rajinder Pal [6]in 2008 further evaluated the several models that had been proposed in the literature to describe the thermal and electrical conductivities of particulate composites. He found that among the proposed models, the Lewis–Nielsen model appears quite attractive as it is simple to use and it predicts the correct behaviour when filler concentration (ϕ) approaches the maximum packing concentration (ϕ_m). In this paper he evaluated the Lewis–Nielsen model in light of a vast amount of experimental data available on thermal and electrical conductivities of particulate composites. The Lewis–Nielsen model was found to describe the experimental data for both thermal and electrical conductivities reasonably well. Zhang et.al.[7] studied the influences of filler size and content on the properties (thermal conductivity, impact strength and tensile strength) of Al_2O_3/high density polyethylene (HDPE) composites. They observed that thermal conductivity and tensile strength of the composites increased with the decrease in particle size. However the dependence of impact strength on the particle size was more complicated. The SEM micrographs of the fracture surface showed that Al_2O_3 with small particle size was generally more efficient for the enhancement of the impact strength, while the 100 nm particles prone to aggregation due to their high surface energy deteriorated the impact strength. Composite filled with Al_2O_3 of 0.5 μm at content of 25 vol% showed the best synthetic properties. They suggested that the addition of nano-Al_2O_3 to HDPE would lead to good performance once suitably dispersed. Nandi et.al. [8] and the others observed that due to poor thermal conductivity of conventional flexible polymeric mould materials, the solidification of (wax/plastic) patterns in soft tooling (ST) process took longer time. This issue could be solved by increasing the effective thermal conductivity of mould materials through (high thermal conductive) particle reinforcement. In this paper, the equivalent thermal conductivities (ETCs) of particle-reinforced polymeric mould materials, namely silicone rubber and polyurethane were experimentally observed using hot disc technique. The findings showed that not only the amount of filler content and type of filler material, but also particle size has significant influence on the effective thermal conductivity of polymer and it starts increasing drastically at 20–30 per cent volume fraction of filler content. To predict the cooling time in ST process, it was important to have an appropriate model of ETC. Here they proposed a new method based on a genetic algorithm fuzzy (GA-fuzzy) approach to model the effective thermal conductivity of a two-phase particle reinforced polymer composites (PCs). The effectiveness of the model was extensively tested in comparison with various empirical expressions reported in literature based on the experimental measurements. It has been found that the model based on GA-fuzzy approach not only outperforms the existing models, but also possesses a generic one applicable to a wide range of two-phase particle-reinforced PCs. Mina et.al. [10] in 2008 used Titanium dioxide (TiO_2) filled isotactic polypropylene with various content of TiO_2. IR spectral studies and X-ray diffraction revealed structural changes from a three-phase (α, β and γ) crystalline system of the neat iPP sample to only a-form due to inclusion of fillers. He observed that Micro-hardness increased rapidly and then levelled off with increasing filler content and also shows variations with varying moulding conditions. Further findings established that the dc electrical resistivity decreased with increasing TiO_2 content and temperature. Lastly he concluded that both the thermal and electrical properties are also found to affect by processing conditions.

Chapter 3

Materials and Methods

3. MATERIALS AND METHODS

The composites would be beryllium oxide particulates in various resins like epoxy, polyester, polypropylene and polyethylene. The high cost of beryllium oxide makes it impractical to get experimental observations, hence effective thermal conductivity would be found out by using the mathematical model and FEM and the results will be compared. The properties of beryllium oxide are listed below.

Properties of BeO (beryllium oxide)

Chemical Formula**:** - BeO

Background: Beryllium oxide is unique because it combines excellent electrically insulating properties with high thermal conductivity. The high toxicity of the powder when inhaled and its high cost have limited its use to application that exploits its singular properties.

Preparation: Beryllium oxide can be prepared by calcining (burning), dehydration beryllium hydroxide or burning the element.

$$BeCO_3 \longrightarrow BeO + CO_2$$

$$Be(OH)_2 \longrightarrow BeO + H_2O$$

$$2\,Be + O_2 \longrightarrow 2BeO$$

In mineral form the most common occurrences of Beryllium are:-

1) Beryl ⟶ Emerald and aquamarine
2) Bertanidite ⟶ Juab county Utah
3) Chrysoberyl ⟶ Alexandrite
4) Euclase ⟶ Topaz

Beryllium is extracted from naturally occurring beryl and bertanadite and produced as a powder by the thermal decomposition of $Be(OH)_2$. Powders are commercially available at purity levels greater than 99%. Components can be made as near net shapes by most of the commonly used fabrication processes like pressing, slip casting or extruding the powder. Sintering is carried out at 1600-1800℃. High density components $< 5\%$ porosity can be easily made with commercially pure powders. Less 1% purity can be achieved using high purity materials and hot pressing in graphite dies.

Physical properties: The physical properties of Beryllium oxide are: -

1) Molar mass-25.01gm/mol
2) Appearance-colourless, vitreous crystal
3) Odourless
4) Density-3.01gm/cm^3
5) Melting point-2507℃
6) Boiling point-3900℃
7) Thermal conductivity-330 W/m K at 0℃
8) Refractive index 1.07
9) Crystal structure-hexagon
10) Young's modulus-345 G Pa
11) Molecular shape-linear
12) Electrical resistivity-$> 1 * 10^{14}$
13) Dielectric strength->9.5kV/mm

Key properties: The key properties are: -

1) Apart from reactivity with water vapours at 1000℃,it is one of the most chemically stable oxides, resisting both carbon reduction and molten metal attack at high temperatures.

2) Thermal conductivity is extremely high in comparison with other ceramics and metals particularly below 300 ℃.

3) Mechanical strength of BeO is lower than alumina but can reach acceptable levels through control of fabrication process.

Methodology: - For the BeO filled composites, the effective thermal conductivity can be found out by using the(Analytical Method) theoretical model for sphere in cube model and the proposed theoretical model derived for cube in cube and hence the results are compared with each other and the results of FEM.

<u>Analytical method</u>

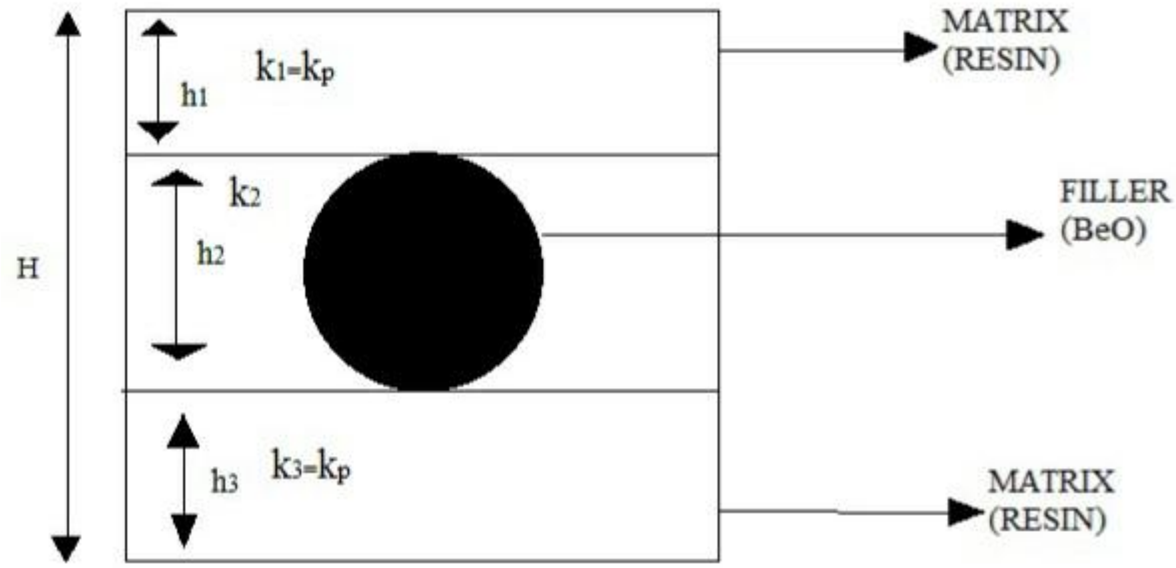

Fig 3.1 Sphere-in-cube-model

The effective thermal conductivity of the sphere in cube model has been derived from the existing sphere in cube model proposed by J.Z. Liang and G.S.Liu [11]. The final expression of this model is

$$k_{\text{eff}} = \frac{1}{\frac{1}{kp} - \frac{1}{kp}\left(\frac{6\Phi}{\pi}\right)^{1/3} + \frac{4}{kp\left(\frac{4\pi}{3\Phi}\right)^{2/3} + (K_f - K_p)\left(\frac{16\pi^2}{9}\right)^{1/3}}}$$

where k_p= Thermal conductivity of epoxy polymer

k_f =Thermal conductivity of filler material

Φ = volume fraction of filler mate

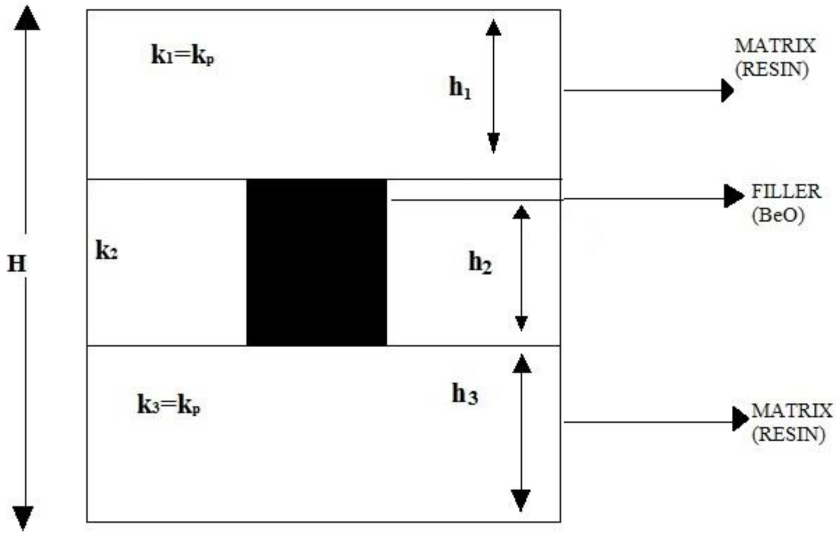

Fig 3.2 Cube-in-cube-model

Here $k_{\text{eff}} = \dfrac{m}{\frac{(m-1)}{k_p}+\frac{m^2}{k_p(m^2-1)+K_f}}$ where $m=(\frac{1}{\Phi})^{\frac{1}{3}}$

Table3.1. Effective thermal conductivity for the various polymers for the theoretical model

ϕ (vol %)	k_{eff} Epoxy (k_p = .363)		k_{eff} Polyester (k_p = .2)		k_{eff} Polypropylene (k_p = .15)		k_{eff} Polyethylene (k_p = .4)	
	sphere	cube	sphere	cube	sphere	cube	sphere	Cube
5	0.667	0.57	0.3677	0.316	0.276	0.237	0.734	0.63
10	0.853	0.68	0.4706	0.372	0.353	0.28	0.939	0.743
15	1.06	0.77	0.585	0.426	0.44	0.32	1.167	0.849
20	1.317	0.87	0.727	0.480	0.546	0.361	1.45	0.96
25	1.64	0.98	0.9135	0.539	0.686	0.404	1.819	1.075
30	2.13	1.09	1.177	0.603	0.884	0.453	2.341	1.203

Numerical Analysis: Concept of Finite Element Method (FEM) and ANSYS:

The Finite Element Method (FEM) was introduced by Turner et al. [58] in 1956, which is a powerful computational technique for approximate solutions for a variety of engineering problems with complex domains subjected to general boundary conditions. FEM has become a crucial step in the modeling of a physical phenomenon in various engineering fields. As the field variables vary from point to point, it results in an infinite number of solutions with in the domain. The basic concept of FEM relies on the decomposition of the domain into a finite number of subdomains (the sample into finite number of elements) for which the systematic approximate solution is constructed by applying either variational or weighted residual methods. FEM reduces the problem into a finite number of unknowns by dividing the domain into elements and expresses the unknown field variable in the form of assumed approximating functions within each element. These functions are also called interpolation functions. These functions define the values of the field variables at specific points called nodes. Nodes connect adjacent elements. This method has the ability to discretize the irregular domains with finite elements for which it is a valuable and practical analysis tool for the solution of boundary, initial and eigen value problems arising in various engineering fields.

Basic Steps in FEM:

The very first step is to convert the governing differential equation into an integral form. The two techniques to achieve this are :

(i) Variational Technique

(ii)Weighted Residual Technique.

In variational technique, the integral form corresponding to the given differential equation is obtained by using calculus of variation. The solution of the problem can be obtained by the minimization of the integral. In weighted residual technique, the weighted integrals of the governing differential equation are constructed where the weight functions are known and arbitrary except that they satisfy boundary conditions.

Often this integral form is modified using the divergence theorem to reduce the continuity requirement of the solution. Then solution is obtained by setting the integral to zero. The second step deals with the division of the domain of the problem into a number of parts, called as elements. This process of division of the domain into a finite number of elements is known as mesh. For one-dimensional (1-D) problems, the elements are nothing but simple line segments having no shape but only length. For problems for higher dimensions, the elements have both the shape and size. For two-dimensional (2-D) or axi-symmetric problems, depending on the type of meshing the elements used are triangles, rectangles and quadrilateral having either straight or curved boundaries. For three-dimensional (3-D) problems, either tetrahedron or parallelepiped elements are used having straight or curved surfaces.

In the third step, for the interpolation functions (also called as shape functions) a proper approximation is chosen as the primary variable and the unknown values of the primary variable at some pre-selected points of the element, called as the nodes. Mostly polynomials are chosen as the shape functions. For 1-D elements, there are at least 2 nodes placed at the endpoints of the line segment. For 2-D and 3-D elements, the nodes are placed at the vertices. Additional nodes are placed on the boundaries or in the interior. Degree of freedom is the value of the primary variable at the nodes.
To get the exact solution, a complete set of polynomials (i.e., infinite term) should be in the expression for the primary variable or if it contains only the finite terms, then the number of elements will be infinite. Each of the above cases results into an infinite set of algebraic equations. Only a finite number of elements and an expression with only finite number of terms are used to make the problem tractable. The accuracy of the approximate solution, however, can be improved either by increasing the number of elements or the number of terms in the approximation.

In the fourth step, the primary variable of approximation is substituted into the integral form. It is minimized to get the algebraic equations for the unknown nodal values if the integral form is of varying type. The algebraic equations are obtained element wise first that is called the element equation and then these element equations are assembled over all the elements to get the algebraic equations for the whole domain (called as the global equations). Then the algebraic equations are modified depending on the boundary conditions and the nodal values are obtained by solving the modified algebraic equations

The last step is the post-processing of the solution. Then, the nodal values are used to construct their graphical variation over the domain either in the form of graphs or contours depending on the dimensions and contours.

Advantages of the finite element method over other numerical methods are as follows:

- Any irregular-shaped domain or any type of boundary condition can be analysed using this method.
- Analysis of domains consisting more than one material can be easily done.
- By proper refinement of the mesh or by choosing higher degree polynomials the accuracy of the solution can be improved.

For the cube-in-cube model a 4×4×4 cm^3 matrix cube is taken and the filler cubes are of size 1×1×1cm^3.

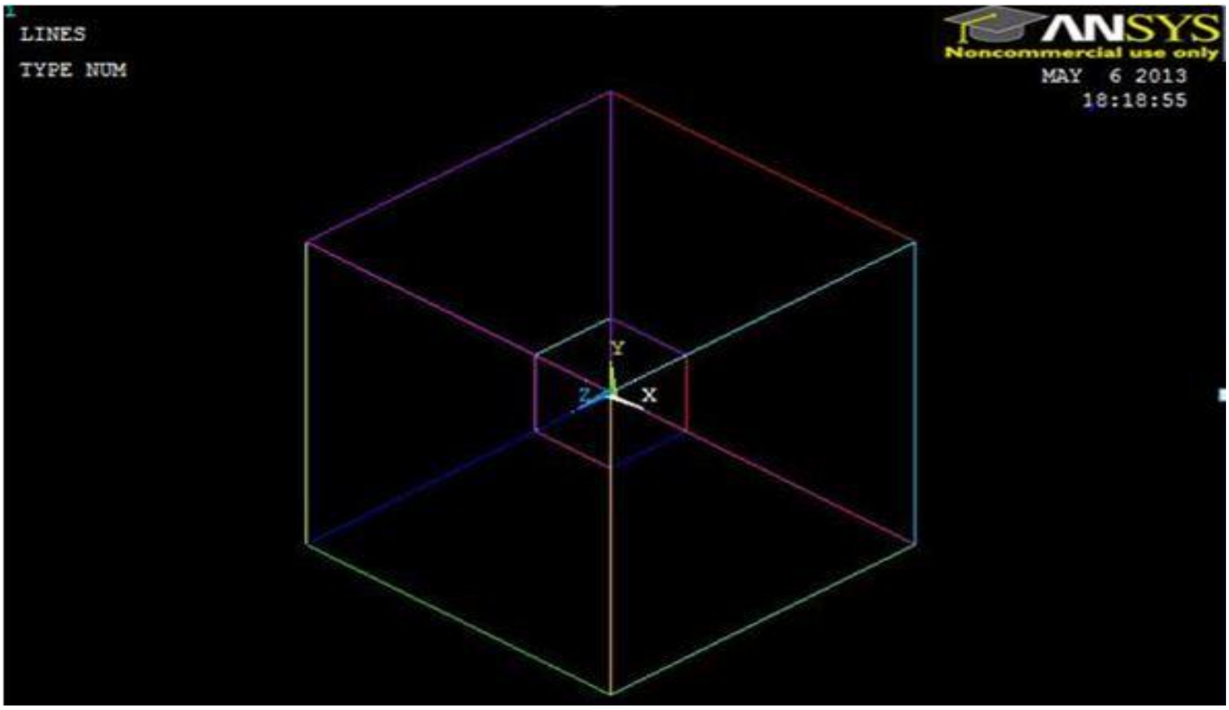

Fig 3.3 One 1×1×1 cube in 4×4×4 cube

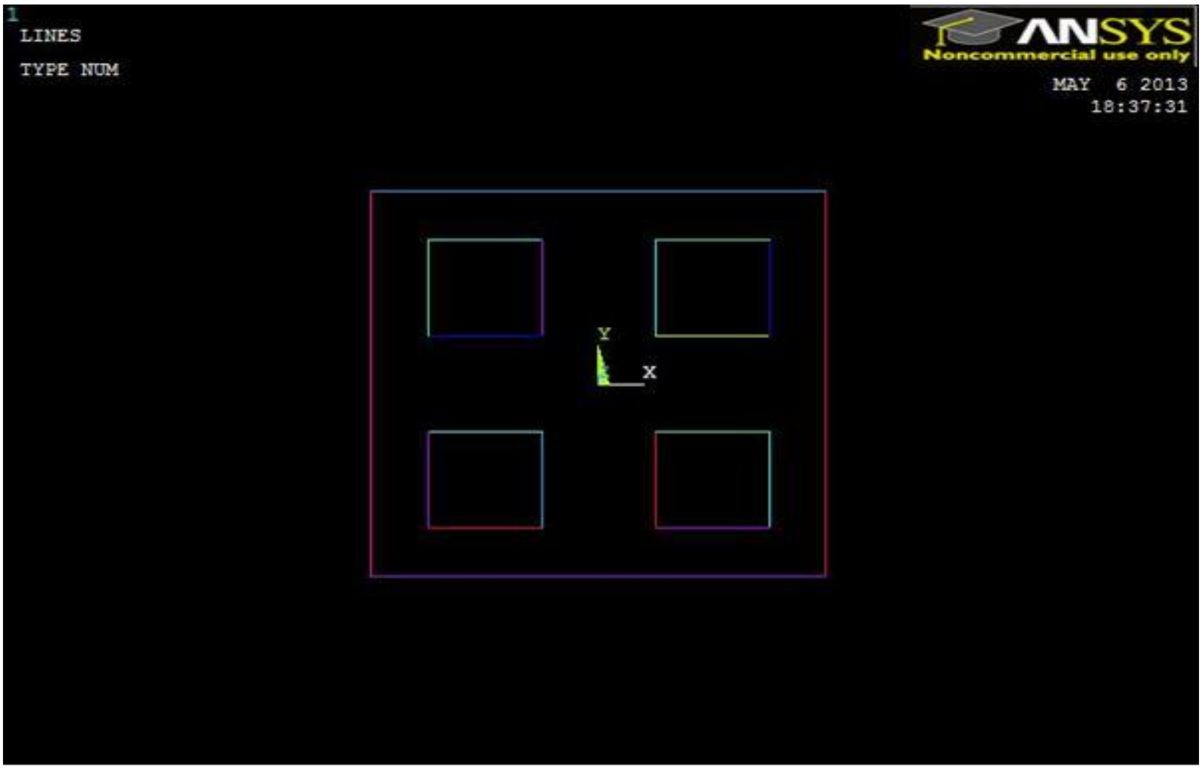

Fig 3.4 Eight 1×1×1 cubes in 4×4×4 cube

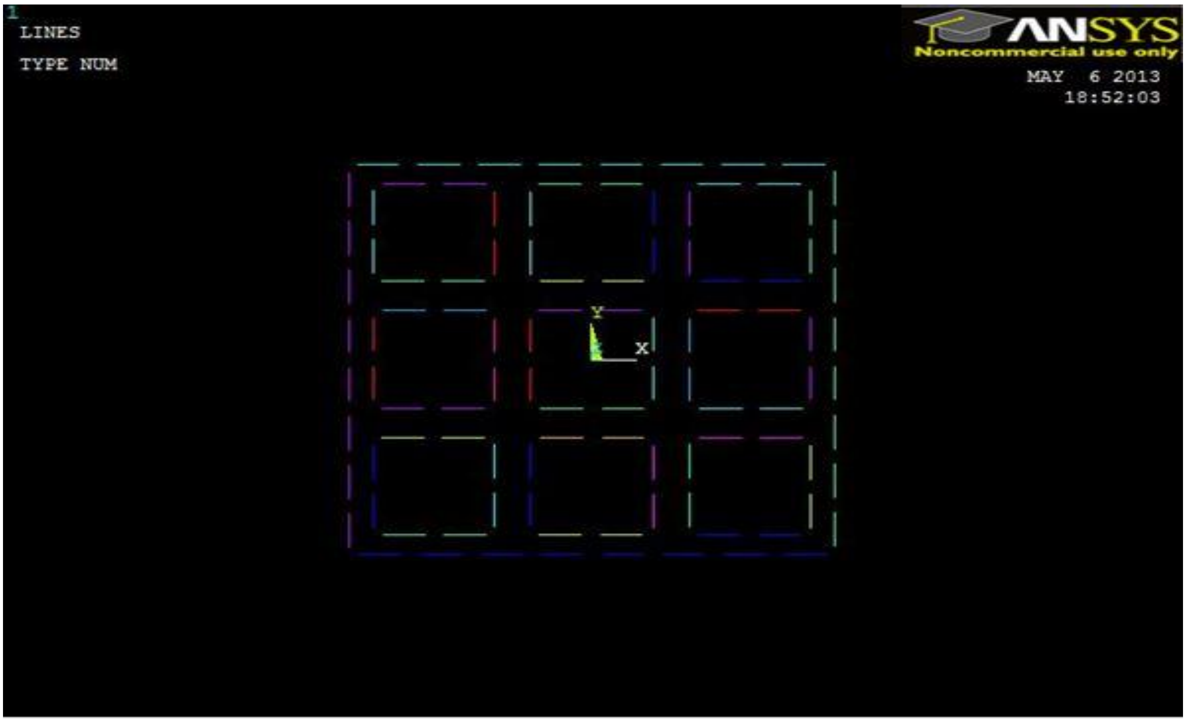

Fig 3.5 Twenty-seven 1×1×1 cubes in 4×4×4 cube

For the sphere-in-cube model a 5×5×5cm^3 cubic matrix is taken and spheres of diameter 1 cm are taken.

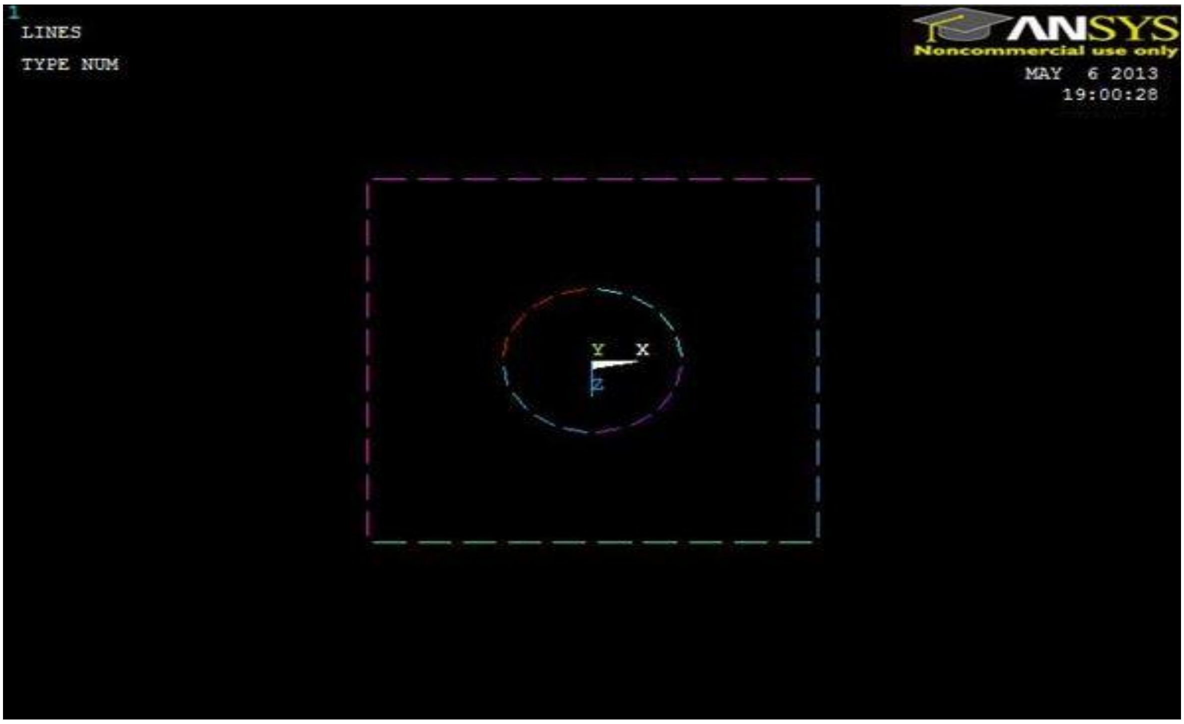

Fig 3.6 1 sphere in 5×5×5 cube

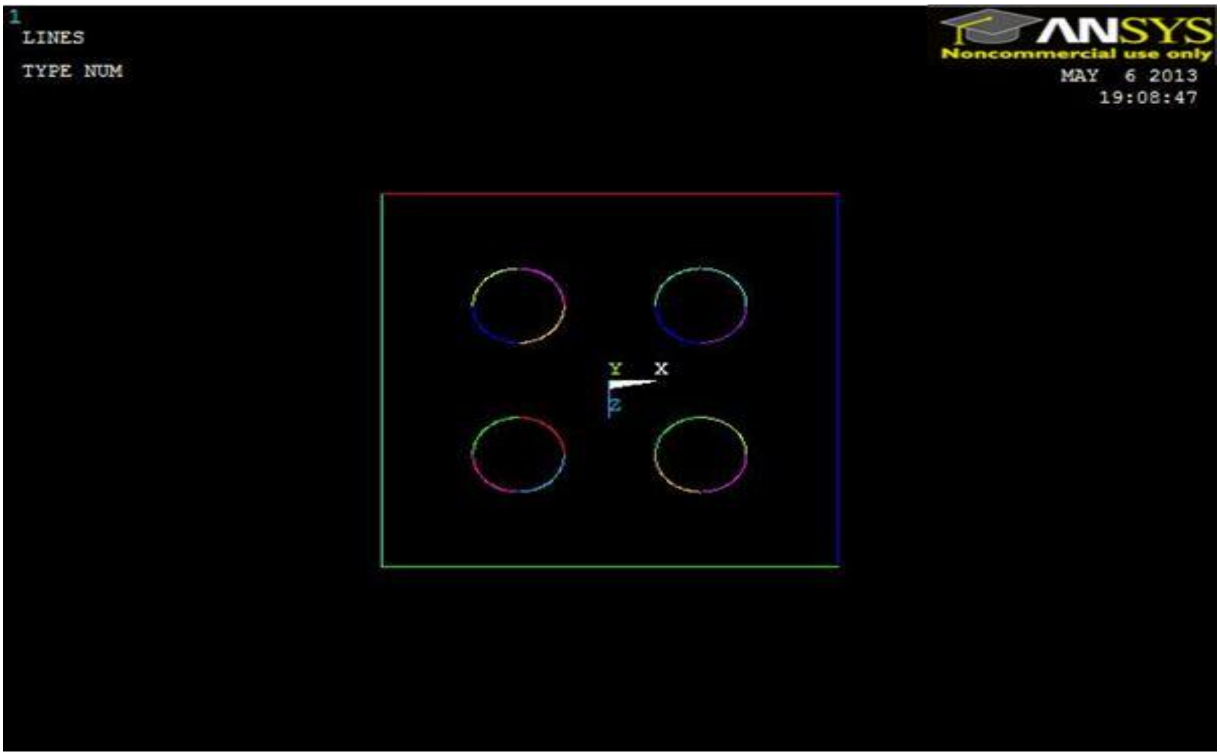

Fig 3.7 8 spheres in 5×5×5 cube

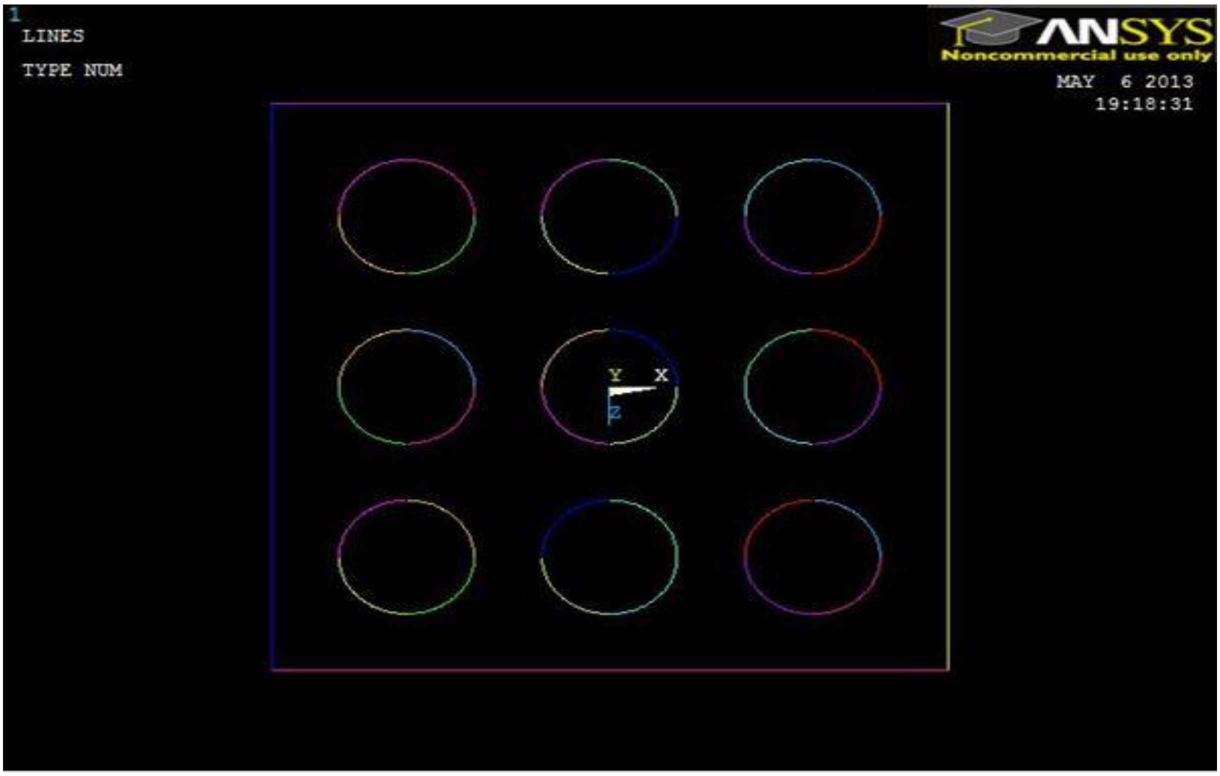

Fig 3.8 27 spheres in 5×5×5 cube

Chapter 4

Results and Discussion

4. RESULTS AND DISCUSSION:

The comparison graph for the various values listed in Table 3.1 is

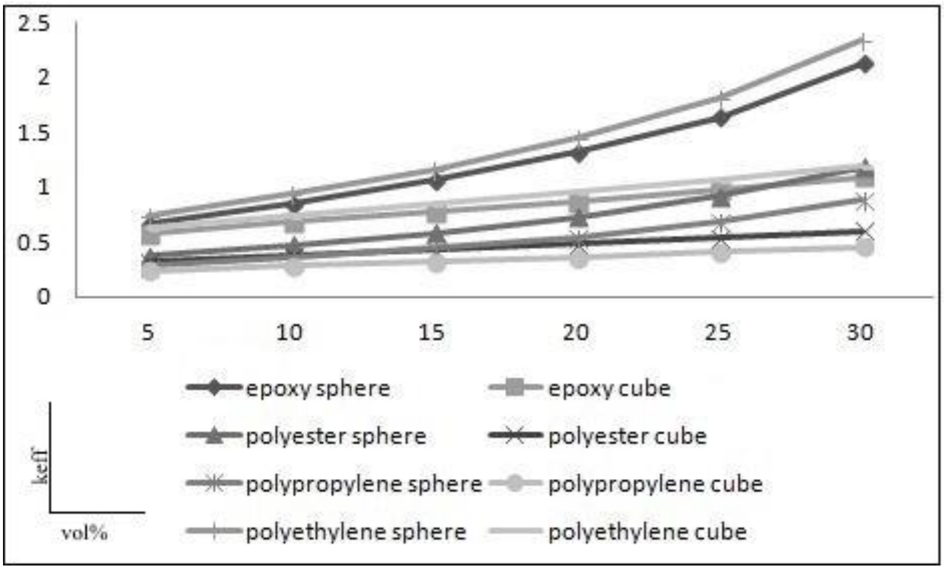

Fig.4.1 Comparison of sphere-in-sphere arrangement for various theoretical values

As we can see from the curves sphere-in-cube model provides higher thermal conductivity than cube-in-cube model and conductivity increase with increase in volume % of filler material. For checking the effect of size we take 20 percent volume fraction and arrange the size of cubes such that the numbers of filler cubes (BeO) are cubic. The temperature profile for the various sizes is plotted with initial temperature 100 °C in each case and final temperatures are compared. A higher temperature at the other end would indicate higher thermal conductivity.

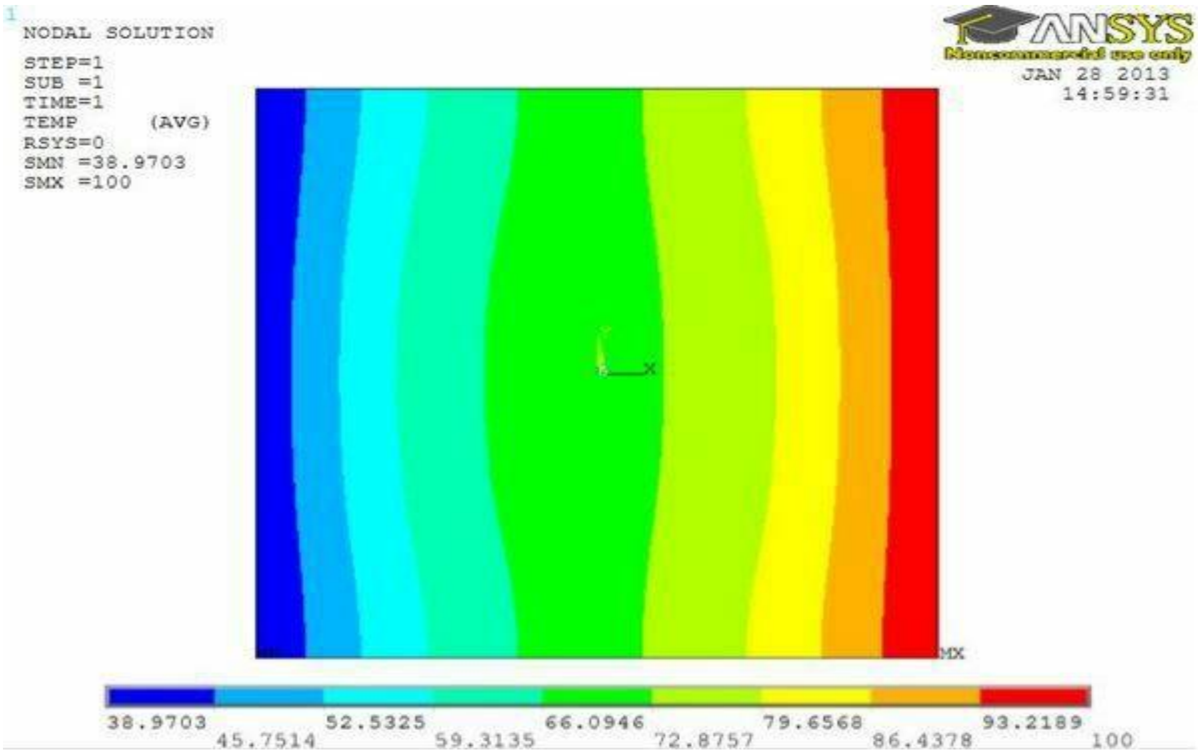

Fig 4.2 For 1 cube at 20 vol %

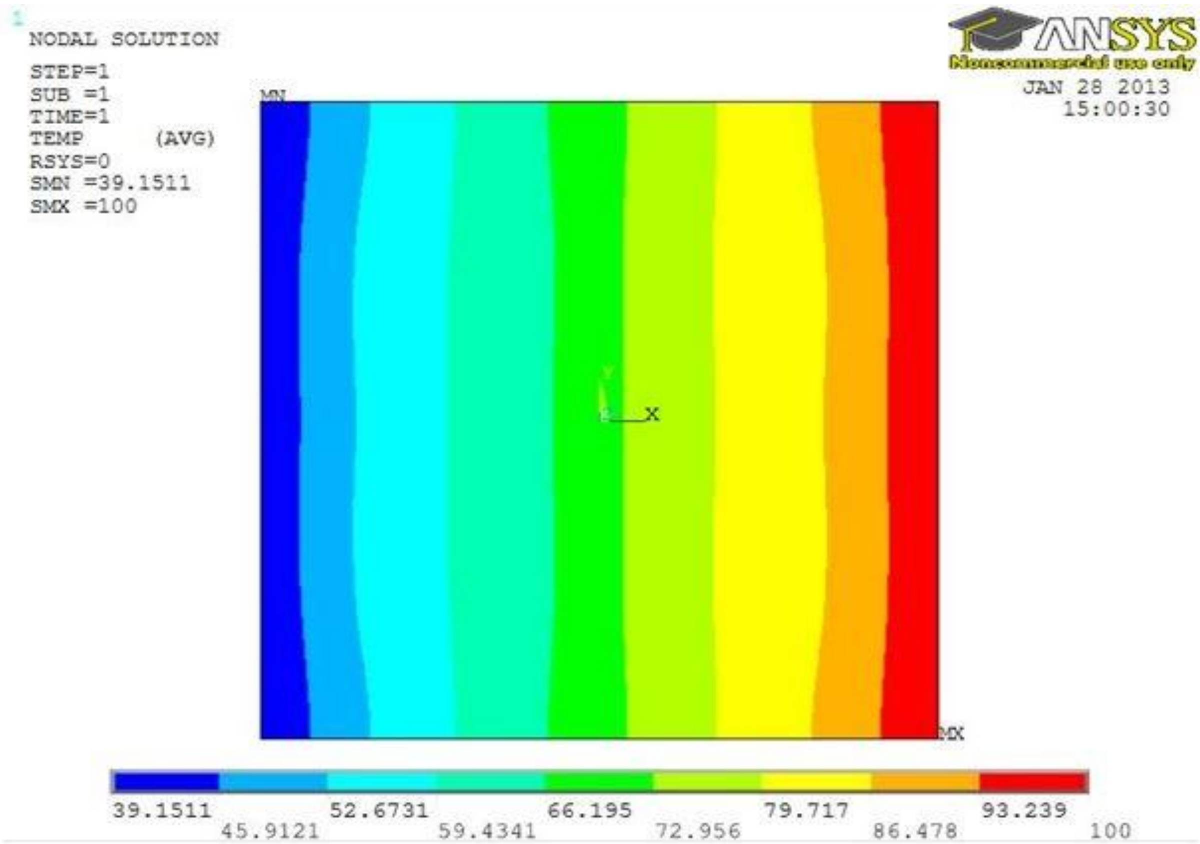

Fig 4.3 For 8 cubes at 20 vol %

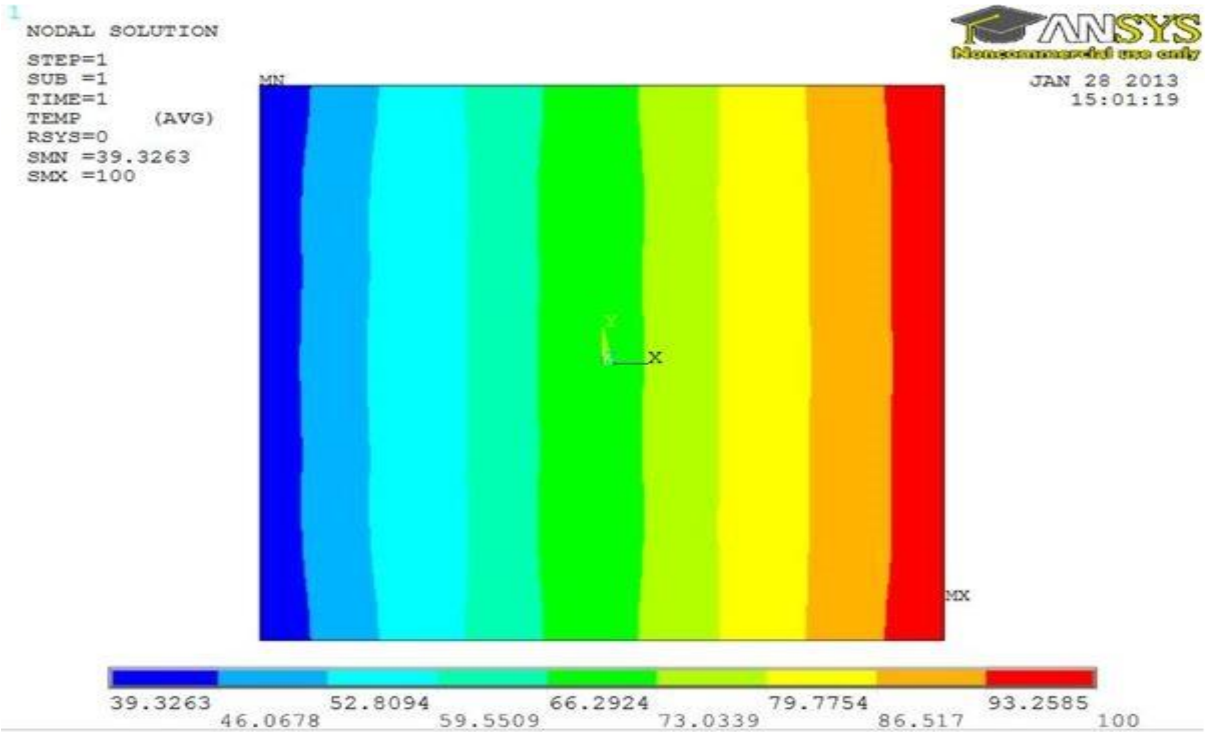

Fig 4.4 For 27 cubes at 20 vol %

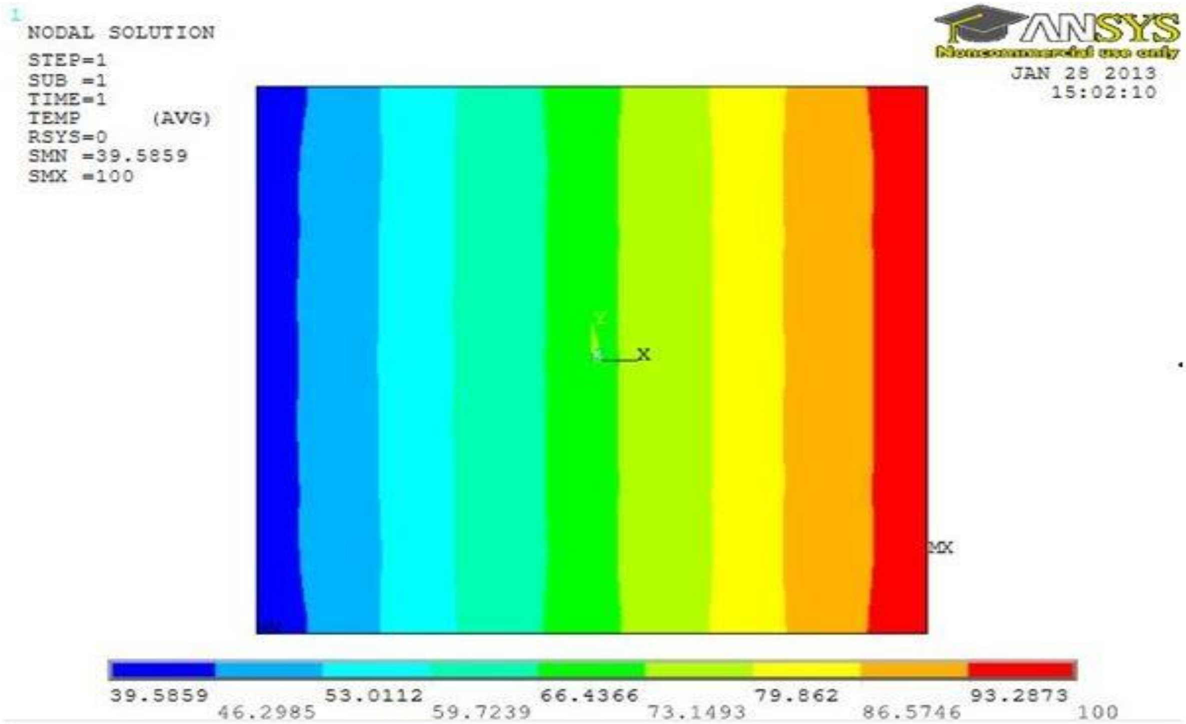

Fig 4.5 For 64 cubes at 20 vol %

Thus we can see that if the number of particles is increased at a particular content % the thermal conductivity increases.

Now in order to compare the results of the mathematical model and FEM we will find the thermal conductivity of 3.3-3.8 from the temperature profile and compare the results to the mathematical model.

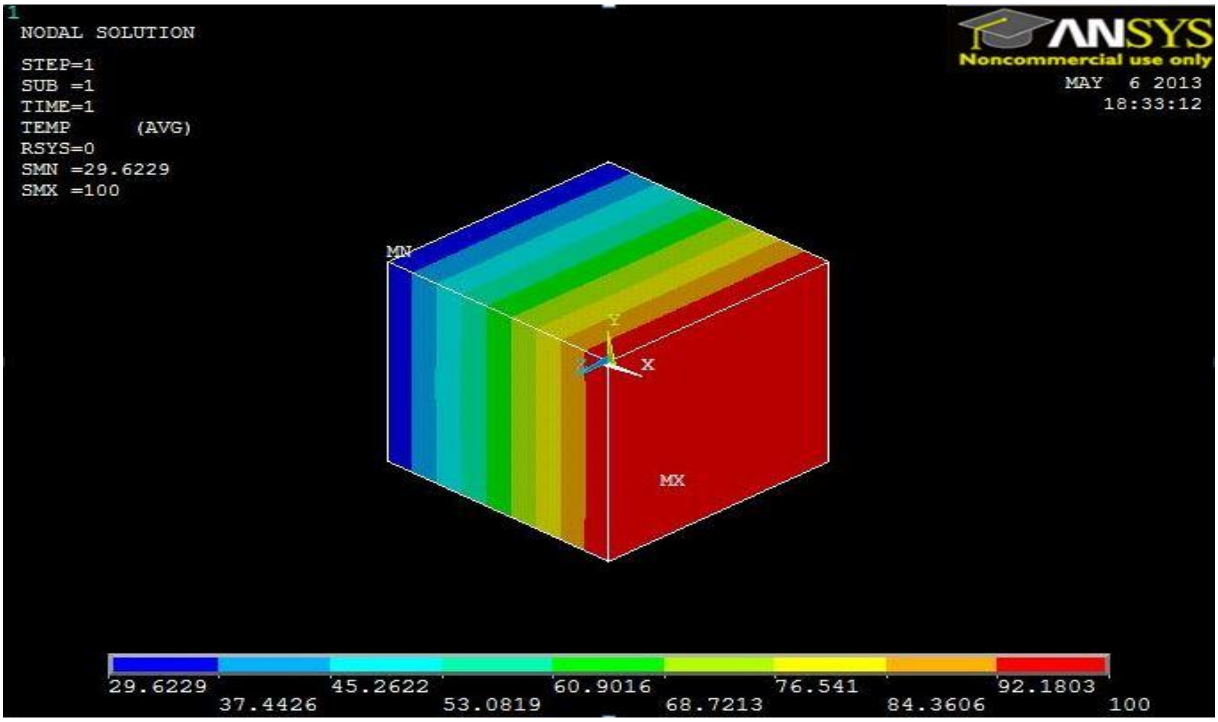

Fig 4.6 Temperature profile for 1 cube-in-cube arrangement

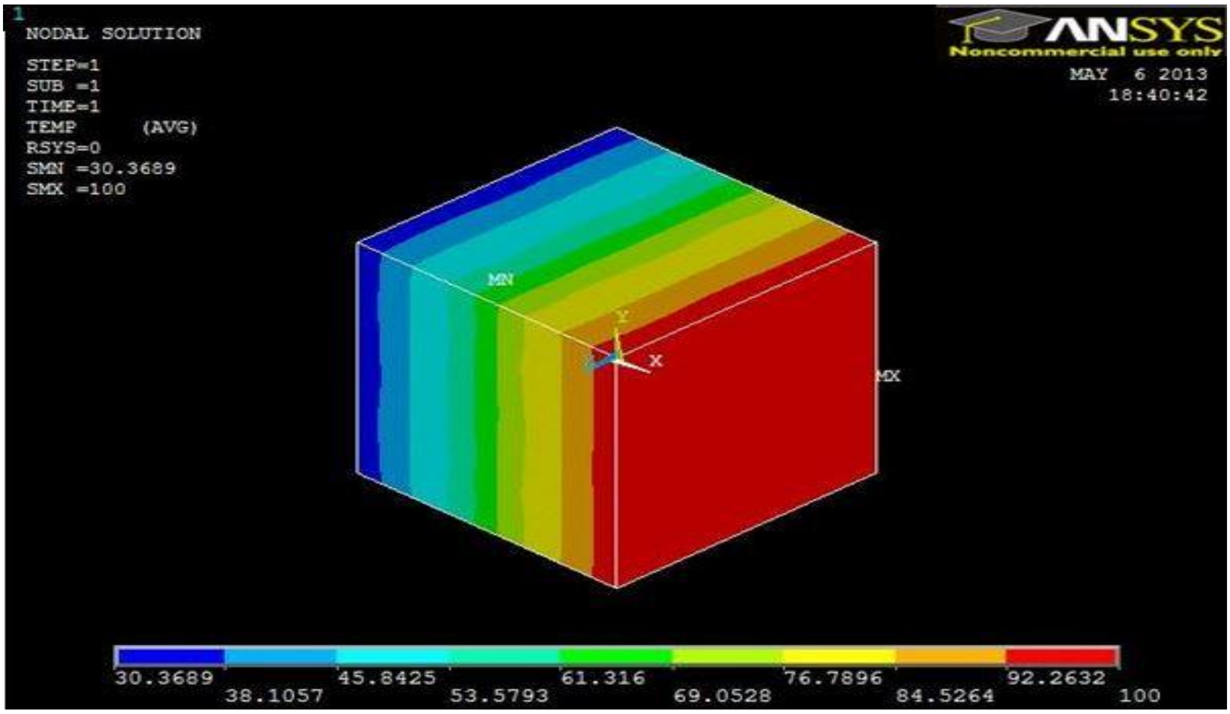

Fig 4.7 Temperature profile for 8 cube -in -cube arrangement

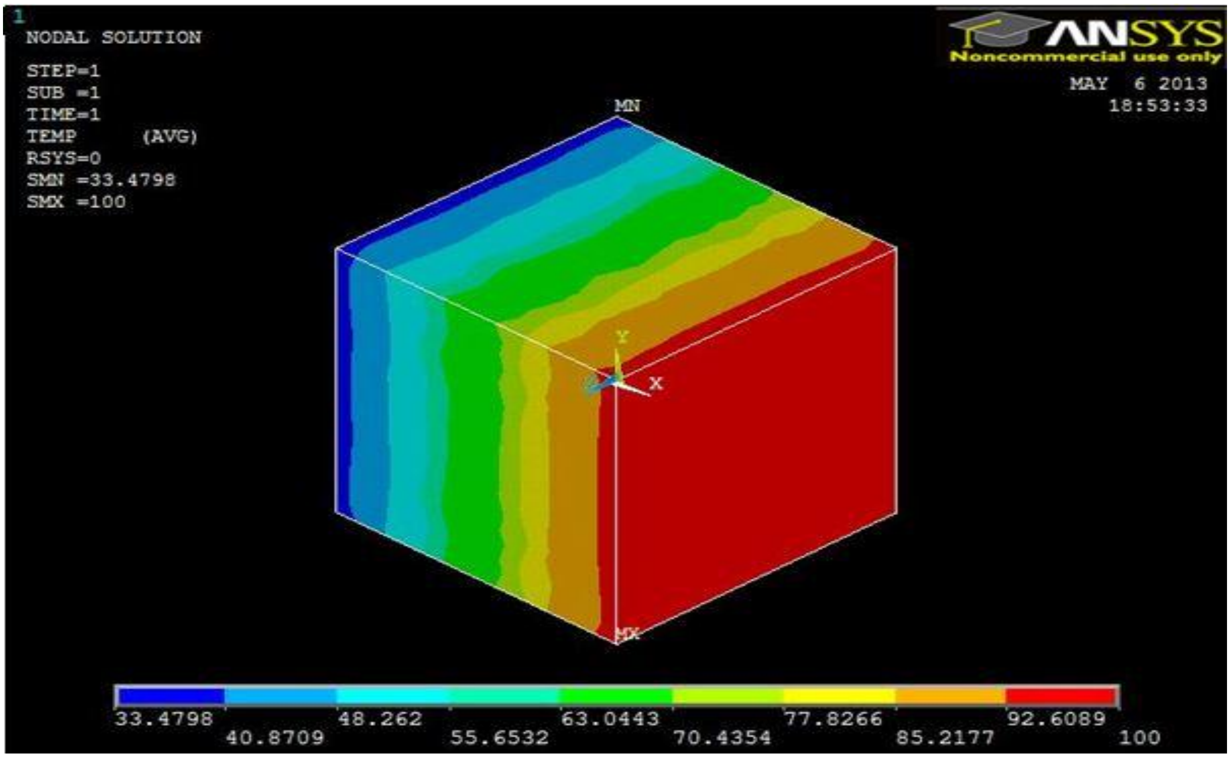

Fig 4.8 Temperature profile for 27 cube - in –cube arrangement

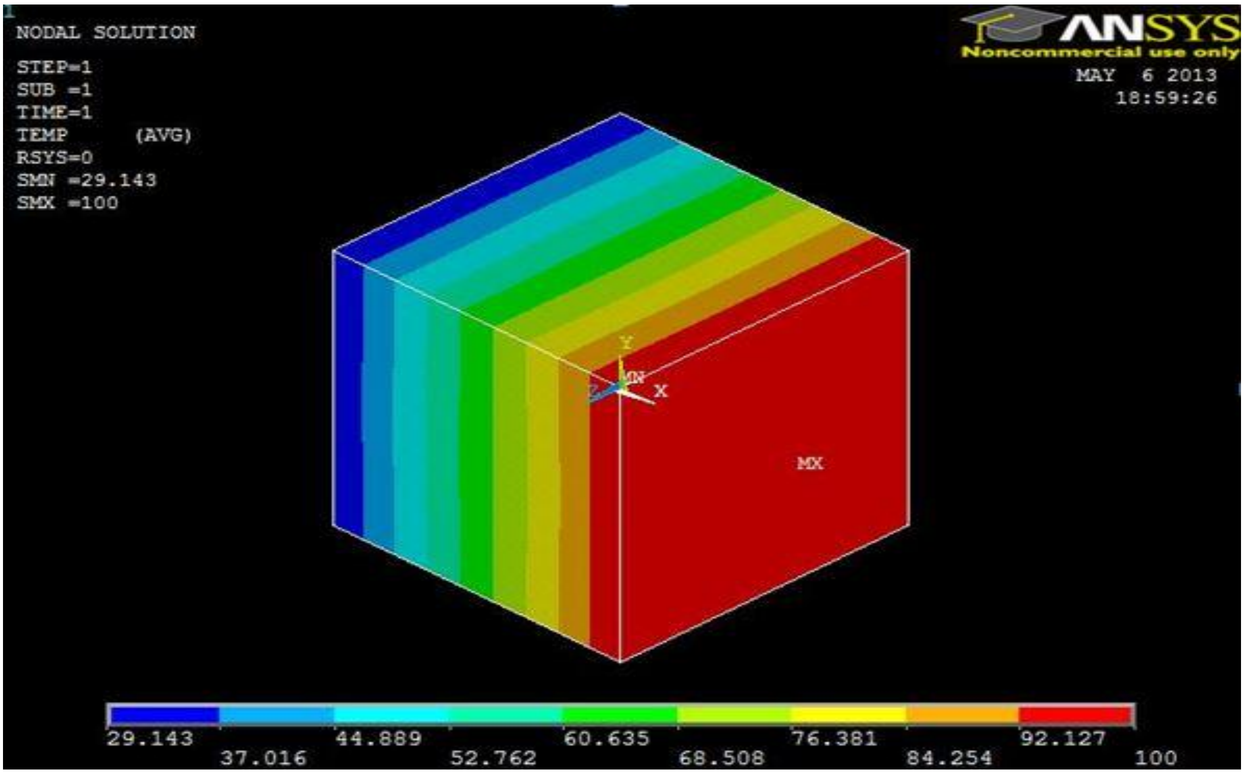

Fig 4.9 Temperature profile for 1 sphere -in -cube arrangement

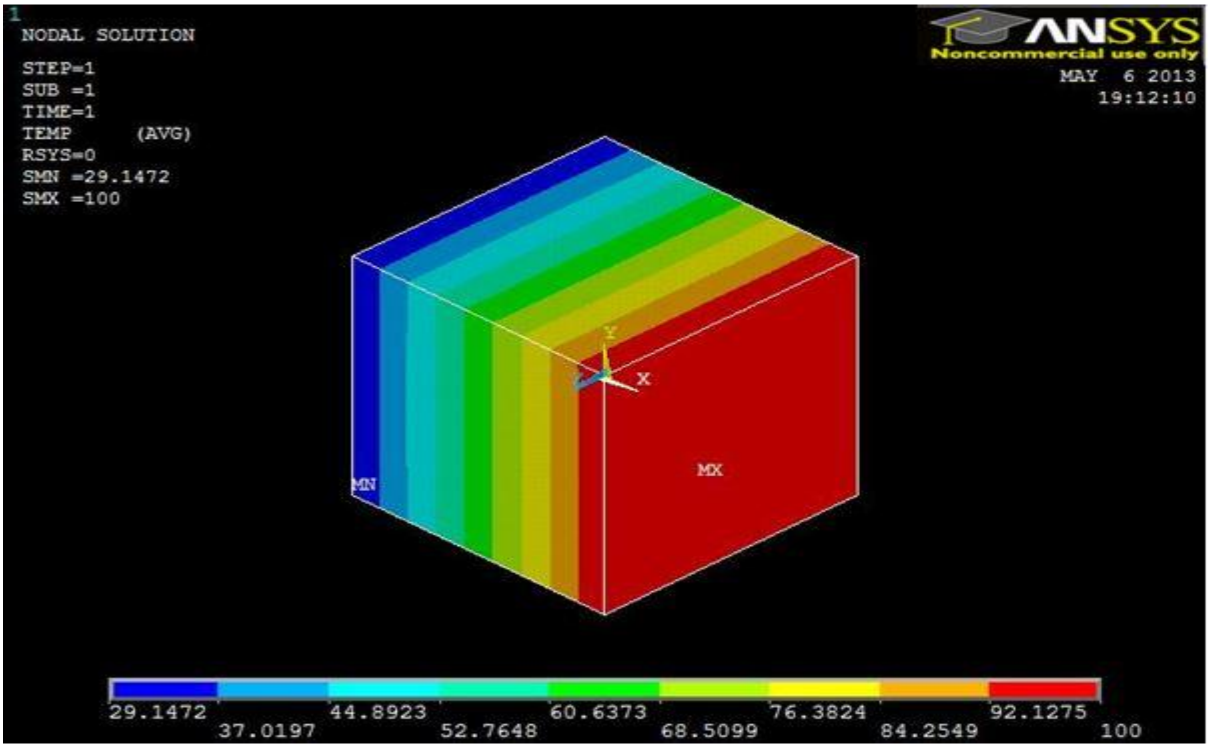

Fig 4.10 Temperature profile for 8 sphere -in -cube arrangement

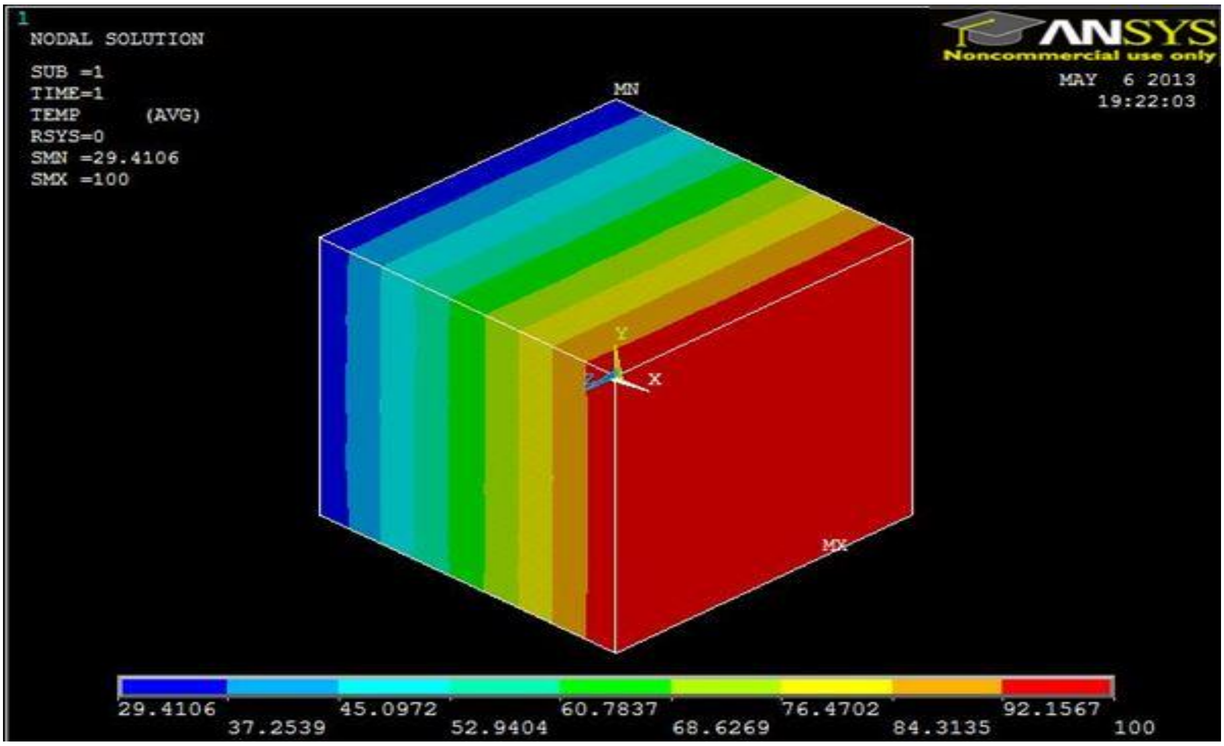

Fig 4.11 Temperature profile for 27 sphere -in -cube arrangement

Table 4.1 Comparision of k_{eff} for FEM and theoretical values for the cube -in -cube model

No of particles	BeO (Vol %)	*keff* (FEM)	*keff* (theoretical model)
1	1.56	0.39	0.48
8	12.5	0.48	0.72
27	42.18	0.98	1.44

Table 4.2 Comparision of k_{eff} for FEM and theoretical values for sphere -in -cube model

No of particles	BeO (Vol %)	*keff* (FEM)	*keff*(theoretical model)
1	0.42	0.37	0.45
8	3.35	0.38	0.61
27	11.3	0.44	0.90

Chapter 5

Conclusions

5.CONCLUSIONS:

1. The effective thermal conductivity of beryllium oxide filled composites increases with increase in filler content.
2. Increase in number of particles increases thermal conductivity.
3. For beryllium oxide, in case of same volume fraction spherical particles have better effect in increasing the thermal conductivity as compared to cubical particles.
4. Finite Element Method (FEM) can be suitably employed to determine the effective thermal conductivity (***keff)*** of these particulate filled polymer composites for different volume fractions of beryllium oxide.

Chapter 6
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